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TRANSFER COEFFICIENTS OF RADIONUCLIDES SECRETED IN THE MILK OF DAIRY COWS

BY DANIEL SAM WALTER F. WILLIAMS
DANIEL D. ROCKMANN JAMES T. ALLEN

RESEARCH AND TECHNOLOGY DEPARTMENT

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Two lactating Holstein cows were administered orally one gelatin capsule per day for 14 consecutive days. Each capsule contained ten radionuclides in their water-soluble forms. The radionuclides were; ^{51}Cr, ^{54}Mn, ^{60}Co, ^{59}Fe, ^{65}Zn, ^{75}Se, ^{125}Sb, ^{131}I, ^{137}Cs, and ^{141}Ce. Milk samples were collected and analyzed in a germanium lithium-drifted detector coupled to a 2048-multichannel gamma-ray analyzer to measure the low levels of complex mixtures of radionuclides. The transfer coefficients for the radionuclides were determined when their secretion in milk reached or approached the concentration plateau.

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SUMMARY

This study was made to simulate experimentally the transfer of radionuclides to the milk of dairy cows on a worst-case situation. Small quantities of the water-soluble forms of ten radionuclides were placed in a gelatin capsule. One capsule per day was administered orally to each cow until each radionuclide secreted in the milk reached or approached the concentration plateau. The radionuclides secreted in cow's milk were measured in a germanium lithium-drifted detector and the transfer coefficient of each radionuclide was determined. This research was conducted to develop information on transfer coefficients of various radionuclides emanating from nuclear power stations and detected on particulates. This information was expected to provide a more reliable basis for constructing dose models for radionuclides emitted on particulates.

This research was sponsored by the Nuclear Regulatory Commission under Contract No. AT(49-24)-0115. The work was performed by the cooperative effort of personnel of the Dairy Science Department of the University of Maryland, College Park, Maryland and the Naval Surface Weapons Center, White Oak Laboratory, Silver Spring, Maryland.

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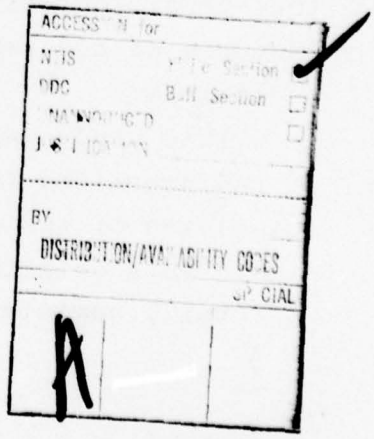

INTRODUCTION

Many radionuclides escaping from nuclear power stations have been identified on particulate matter. It has been shown that these particulates fall out on adjacent pastures and could be assimilated by grazing cattle and thereby secreted into the milk of dairy cows. Since milk and milk-products were widely used as food sources, studies of dairy cows designed to measure the milk transfer of various radionuclides provide important information for estimating the potential hazard from the milk link in man's nutrient chain.

Some of the radionuclides found to be emitted on particulates were: ^{51}Cr, ^{54}Mn, ^{60}Co, ^{59}Fe, ^{65}Zn, ^{75}Se, ^{124}Sb, ^{131}I, ^{137}Cs, and ^{141}Ce. These radioisotopes were used to simulate secretion in the milk of dairy cows under conditions of continuous intake at a constant level. However, ^{125}Sb was used in place of ^{124}Sb due to the presence of interfering gamma-rays in the complex spectrum of the latter.

The above radioisotopes were most convenient for this study because their half lives were sufficiently long compared to the lengths of the experiments, their numerous gamma-ray energies were easily separable on a germanium lithium-drifted detector and their specific activities were either carrier-free or sufficiently high so that physiological interferences due to the presence of stable isotopes of the same elements in the same chemical species were negligible.

This study was conducted to simulate experimentally the transfer of radionuclides to cow's milk on a worst-case situation. Small quantities of the water-soluble form of each radionuclide were placed in a gelatin capsule. One capsule containing ten radionuclides was orally administered daily to each cow until each radionuclide

secreted in the milk reached or approached the concentration plateau. Oral administrations of gelatin capsules were continued for 14 consecutive days. During the dosing period, the dairy cows were fed on grass, hay, or green chop, a feeding regimen widely used in the Eastern United States.

The transfer of radionuclides to cow's milk was studied with a germanium lithium-drifted detector coupled to a 2048-multichannel gamma-ray analyzer. The detector with its high resolution was most convenient for detection and separation of individual radionuclides in samples containing low levels of complex mixtures of numerous radionuclides. The radionuclides secreted in the milk were identified and specific radionuclides were quantitatively determined from the prominent photopeak areas of the corresponding gamma-ray spectrum.

Since the duration for transfer of a metabolite to milk was relatively short, in the order of a few days, it was expected that nearly all radionuclides under investigation would have reached or approached the concentration plateau during the dosing period. However, for some radionuclides, a gradual but small rise in milk transfer could continue for longer periods. Nevertheless, this rise was expected to be negligibly small.

EXPERIMENTAL

Two lactating Holstein cows were selected for this study. The cows weighed 611.3 and 577.2 kg, respectively. The average daily milk production was 18.9 and 20.7 kg, respectively. Throughout the study, the cows were subjected to identical experimental conditions. After the cows had been catheterized, they were allowed two weeks to become accustomed to the limited confinement of the metabolism stalls, milking time, and feeding schedules. Then samples of milk, feces , urine, and blood were taken for background activity measurement.

Immediately following the pre-treatment period, one gelatin capsule containing ten radionuclides was orally administered daily to each cow for 14 consecutive days. An initial estimate of the daily required dose of each radionuclide was made so that the daily dose administered to each cow was not so large that undesirable physiological effects were induced. The daily administered dose for each radionuclide was between 10 and 100 microcuries.

The gelatin capsules were prepared in the following manner. Ten radionuclides in various chemical forms, as shown in Table 1, were placed in gelatin capsules. The radionuclide solutions were obtained commercially. Each solution was diluted with the desired volume of acid or alkali to make up to a convenient stock solution. A known aliquot of each stock solution was introduced into the gelatin capsules. To avoid possible dissolution of the gelatin capsules, the stock solutions were deposited on finely grounded dried hay particles which were used to fill the capsules.

It was also necessary to separate the acidic solutions from the alkaline iodide solution, because iodide may be converted to

TABLE 1

CHEMICAL FORMS AND SPECIFIC ACTIVITIES OF VARIOUS RADIONUCLIDES FOR ORAL ADMINISTRATION TO DAIRY COWS

Radionuclides	Chemical Form	Specific Activity	Total Solids
^{141}Ce	$Ce(NO_3)_3$ in 0.5 N HNO_3	0.495 mCi/mg	10 mg/ml
^{75}Se	H_2SeO_3 in 0.5 N HCl	134 mCi/mg	0.15 mg/ml
^{51}Cr	$CrCl_3$ in 0.5 N HCl	104 mCi/mg	0.22 mg/ml
^{131}I	NaI in 0.05 N NaOH	Carrier-Free	2 mg NaOH/ml
^{125}Sb	$SbCl_3$ in 4.0 N HCl	Carrier-Free	$<$0.1 mg/ml
^{137}Cs	CsCl in 0.5 N HCl	Carrier-Free	$<$0.1 mg/ml
^{54}Mn	$MnCl_2$ in 0.5 N HCl	Carrier-Free	$<$0.1 mg/ml
^{59}Fe	$FeCl_3$ in 0.5 N HCl	23.6 mCi/mg	0.92 mg/ml
^{65}Zn	$ZnCl_2$ in 0.5 N HCl	Carrier-Free	$<$0.1 mg/ml
^{60}Co	$CoCl_2$ in 0.5 N HCl	80.6 mCi/mg	0.27 mg/ml

NOTE: (1) The radiometric purity of each radionuclide is 99% or greater.

elemental iodine in acidic solution resulting in possible loss of radioiodine through sublimation. Therefore, the acidic radionuclide solutions were placed in a small gelatin capsule of 1.42 cm^3, while the alkaline radioiodine solution was placed in another gelatin capsule of similar size. These two small capsules were then placed inside a large gelatin capsule of 14.2 cm^3. Double encapsulation was used to minimize the possibility of capsule rupture during dosing.

The gelatin capsules were administered orally using a balling gun. During this period, feed consumption of each cow was measured. The cows were maintained on their regular ration of 18 and 19.3 kg of fresh grass, hay and green chop, respectively. A grain mix was fed to the cows twice daily at milking time, based on body weight and milk production. The grain mix consumption was 9 and 11 kg, respectively. The cows were allowed free access to water.

Milk, urine, and feces production were measured daily and until the time of slaughter. The cows were slaughtered at about 37 days after end of radionuclide administration. All samples obtained after slaughter were weighed, and estimates of the total weights were calculated for muscles, blood, and edible organs.

During the course of the experiment, the cows contracted acute mastitis. Treatment was initiated before the cows showed a decrease in feed consumption, milk production, or a fever. Samples were taken and counted regardless of the presence of mastitis.

A constant sample volume of 60 cm^3 was taken for measurement to assure a reproducible counting geometry. The sample container was a wide-mouth polyethylene bottle of 70 cm^3 and was provided with a tightly sealed screw cap. The samples were frozen and maintained at -10°C to -29°C until measurement. All radioactivity measurements were made in a Ge(Li) detector coupled to a Nuclear Data 2048-multichannel gamma-ray analyzer. The 2048-multichannel gamma-ray analyzer was calibrated at 0.72 Kev per channel to cover the gamma-ray energy range of the ten radionuclides under study. The energy calibration was made by counting a composite radionuclide source consisting of ^{22}Na, ^{241}Am, ^{137}Cs, and ^{60}Co.

The half-lives and the gamma-ray energies of these radionuclides are given in Table 2. The gamma-ray analyzer was deliberately offset to cut out the first 20 channels from the spectrum to avoid overloading effects.

TABLE 2

COMPOSITE RADIOACTIVE SOURCE USED FOR ENERGY CALIBRATION OF THE 2048-MULTICHANNEL GAMMA-RAY ANALYZER

Radionuclides	Half-Lives	Gamma-Ray Energies (MeV)
^{241}Am	433 yrs	0.060
^{137}Cs	30.2 yrs	0.662
^{22}Na	2.602 yrs	1.274
^{60}Co	5.258 yrs	1.173 1.330

The Ge(Li) detector was 30 cm^3 and it was cooled in liquid nitrogen. The detector was right-cylindrically shaped with its axis of symmetry horizontal, so that the sample containers were positioned with their side facing toward the flat surface of the detector. The detector was shielded on all sides with at least 3 inches of lead bricks. For counting, the samples were placed in a special polyethylene holder which was designed to assure a reproducible geometry with respect to the detector. The gamma-ray spectrum of each sample was displayed on an oscilloscope and the digital data were printed out by a teletype machine.

All samples were counted, without further processing, on the Ge(Li) detector. Aliquots of standard radionuclide stock solutions for oral administration to the cows were measured in the same geometry as the milk samples. The integrated counts in the major photopeaks of each radionuclide, less background, were used to determine the concentration of each radionuclide in the sample.

DATA REDUCTION OF A TYPICAL PHOTOPEAK

The digital data contained in each photopeak of interest were plotted and a smooth curve was drawn through the points. The peak-height for each channel of the photopeak was taken from the smooth curve. The photopeak area, which was directly proportional to the quantity of radioactivity in the sample was determined according to the method reported by Covell[1]. This method is briefly described below.

Figure 1 shows a schematic sketch of a typical photopeak presented as a bar graph. Here, the channel containing the greatest number of counts was defined as a_o and succeeding channel responses progressing down the low-amplitude side of the peak as a_1, a_2, a_3, ...a_n. Similarly, channel responses on the high-amplitude side of the peak were defined as b_1, b_2, b_3, ...b_n.

If the contributions from the a_o channel and the n channels on either side were added, the sum, P, represented the total counts contained in these channels.

$$P = a_o + \sum_{1}^{n} a_i + \sum_{1}^{n} b_i$$

The partial area, N, bore a constant relationship to the total area contained in the peak so that N was used to estimate the total peak area or more directly the gamma-ray abundance, which was directly proportional to the concentration of the radionuclide in the sample. The remaining partial area was called Q.

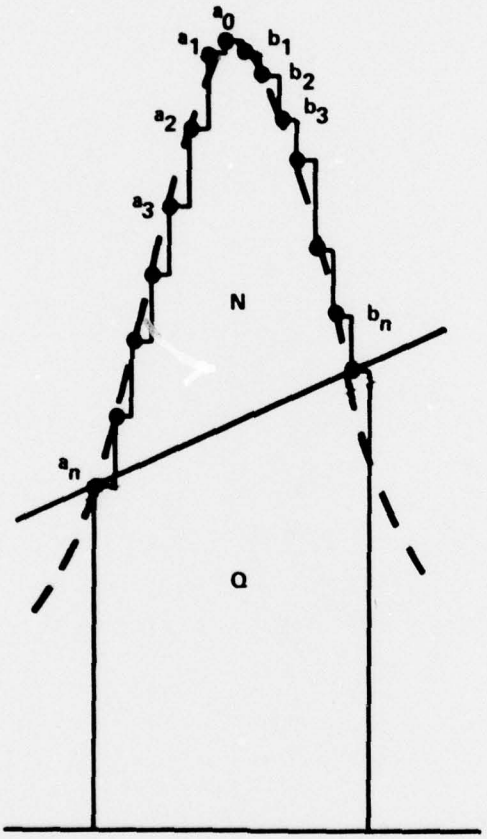

FIGURE 1 SCHEMATIC SKETCH OF A TYPICAL PHOTOPEAK

$$N = P - Q$$

$$N = a_o + \sum_{1}^{n} a_i + \sum_{1}^{n} b_i - (n + \tfrac{1}{2})\ (a_n + b_n)$$

For convenience in the estimation of the photopeak areas, we chose, arbitrarily, n = 10 for photopeaks of ^{60}Co and n = 6 for the photopeaks of the other radionuclides.

RESULTS AND DISCUSSION

The experimental results of the transfer coefficient of ten radionuclides secreted in the milk of two lactating Holstein cows were given in Table 3. All radiological data were corrected for decay and aliquot size with the zero time taken to be the time of initial radionuclide administration. Only milk transfer data taken from 10 to 14 days after initial administration were presented because we were reporting mainly the transfer coefficients as the radionuclides secreted in milk reached or approached the concentration plateau.

Transfer coefficients were expressed as the percent of the daily administered dose secreted per kilogram of milk. Our transfer coefficients were expressed by weight, while those given in the literature were usually expressed by volume. Since the density of milk was nearly unity, the transfer coefficients, whether expressed by weight or by volume, for all practical purposes, were directly comparable.

Referring to Table 3, the daily transfer coefficients of each radionuclide secreted in milk remained roughly constant. During the 10 to 14 days of dosing, variations occurred in amounts secreted in milk presumably as the result of stress factors caused by prolonged confinement in the metabolism stalls.

The daily transfer coefficients of each cow gave fair agreement for some radionuclides. The average transfer coefficients for ^{131}I, ^{137}Cs, ^{54}Mn, ^{60}Co were 0.88%, 0.79%, 0.033%, and 0.01%/kg of milk, respectively. These values were in good agreement with literature values for ^{131}I and ^{137}Cs.

The recorded transfer coefficients for ^{131}I ranged from 0.2% to 2.7%/ℓ[2,3,4] of milk with 90% of these values occurring within

TABLE 3. TRANSFER COEFFICIENTS OF RADIONUCLIDES IN COW'S MILK AT VARIOUS TIMES AFTER INITIAL ADMINISTRATION

Transfer Coefficients (%/kg of milk)													
Radio-nuclides	Cow #1 Time After Initial Administration (Days)					Average		Cow #2 Time After Initial Admin.		Average		Average (For 2 Cows)	
	10	11	12	13	14			12	14				
^{131}I	0.66	0.75	0.65	0.58	0.67	0.66	±9.2%	1.1	1.1	1.1		0.88	±6.2%
^{137}Cs	0.77	0.75	0.74	0.58	0.81	0.73	±12%	0.80	0.87	0.84	±5.9%	0.79	±10%
^{54}Mn	0.029	0.036	0.035	0.026	0.026	0.030	±16%	0.038	0.032	0.035	±12%	0.033	±15%
^{60}Co	0.012	0.0091	0.014	0.0081	0.011	0.011	±20%	0.0092	0.0082	0.0087	±8.1%	0.01	±22%
^{125}Sb		0.014	0.0085	0.015	0.014	0.013	±23%	0.0082	0.0089	0.0086	±5.8%	0.011	±24
^{65}Zn	0.39	0.42	0.40	0.29	0.48	0.40	±17%	0.24	0.19	0.22	±16%	0.31	±21%
^{75}Se	0.34	0.35	0.33	0.57	0.37	0.39	±26%	0.13	0.24	0.19	±42%	0.29	±33%
^{59}Fe	0.0058	0.0064	0.010	0.0044	0.0079	0.0069	±31%	0.0024	0.0028	0.0026	±11%	0.0048	±40%
^{141}Ce								0.0078	0.018	0.013	±56%	0.013	±56%
^{51}Cr												(0.01)	

NOTE: (1) Standard deviation is expressed in percent of the average value.

(2) ^{141}Ce was too low for detection in milk samples of cow #1.

(3) Transfer coefficient written in parenthesis is an estimated upper-limit. ^{51}Cr was too low for detection in milk samples of both cows.

the range of 0.5% to 2.0%/ℓ. Therefore, Tamplin's[2] recommendation of 1%/ℓ of the total daily ^{131}I intake appeared in milk was not unreasonable as an average transfer coefficient expected under conditions of continuous ^{131}I deposition. Our experimental transfer coefficient for ^{131}I is 0.88%/kg of milk.

The uptake of ^{137}Cs in milk has been studied extensively under laboratory and field conditions. Kahn[5] et al studied the transfer of ^{137}Cs to milk under field conditions. They found that on three feeding regimens, free grazing, stored corn silage, or recently harvested grass silage, all supplemented with grain approximately 0.7%/ℓ of ^{137}Cs ingested daily was transferred to milk. Pelletier and Voilleque[6] found that 0.46%/ℓ of the daily ^{137}Cs intake from fallout appeared in milk. Ward and Johnson[7] found that the ^{137}Cs content for each liter of milk was 0.24% and 0.58%/ℓ for high-hay and high-grain diets, respectively. Our experimental transfer coefficient for ^{137}Cs was 0.79%/kg of milk.

The transfer coefficient of ^{54}Mn gave good agreement between the two cows. Our average experimental transfer coefficient for ^{54}Mn was 0.033%/kg of milk. Ng[8] et al compiled a comprehensive report on the transfer coefficients of various radionuclides derived from numerous available tracer studies based on an extensive literature review. Ng[8] et al reported an estimated value of <0.0001%/ℓ for ^{54}Mn, while Pelletier and Voilleque[6] estimated that <0.049%/ℓ of the ^{54}Mn daily intake appeared in milk. There existed a wide discrepancy between the former transfer coefficient value and the present experimental value.

According to Table 3, the average transfer coefficients between the two cows for ^{65}Zn, ^{59}Fe, and ^{75}Se differed by about a factor of two or greater. However, the average transfer coefficients of both cows for these radionuclides were 0.31%, 0.0048%, and 0.29%/kg, respectively. Ng[8] et al gave estimated values for ^{65}Zn, and ^{59}Fe as 0.32%/ℓ and 0.0002%/ℓ, respectively. Our experimental value for ^{65}Zn gave exceptionally good agreement with this reported value. Whereas the estimated value for ^{59}Fe was smaller than our experimental value by a factor of about 25.

Ng[8] et al reported estimated values of 0.12% and 0.37%/ℓ for ^{75}Se from two different sources. Our experimental value for ^{75}Se lay between these estimates.

The average transfer coefficients for ^{60}Co and ^{125}Sb between the two cows were in rough agreement. Our average transfer coefficients for ^{60}Co and ^{125}Sb were 0.01% and 0.011%/kg, respectively. For a different radioantimony nuclide, Ng[8] et al reported the value of 0.002/ℓ for ^{124}Sb. This estimated value of ^{124}Sb was smaller than our experimental value for ^{125}Sb by a factor of about 5.

The ^{141}Ce radionuclide was barely detected in the milk of one of the cows. Our experimental transfer coefficent for ^{141}Ce was 0.013%/kg of milk. Ng[8] et al gave an estimated value of 0.005%/ℓ However, for a different radiocerium nuclide, Garner[9,10] et al reported that the transfer coefficient of ^{144}Ce was about 0.01%/ℓ.

The ^{51}Cr radionuclide was too low for detection by the gamma-ray analyzer. The maximum transfer coefficient for this radionuclide, estimated from the minimum detectable quantity, was 0.01%/kg.

The present data showed that the experimental transfer coefficients for radionuclides secreted in the milk of dairy cows fed on grass, hay or green chop, a feeding regimen used in the Eastern U. S., were in general agreement with reported transfer coefficients for ^{131}I, ^{137}Cs, and ^{65}Zn. However, our data indicated marked difference in transfer to milk from that reported in the literature for ^{59}Fe.

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